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$^{233}U/^{236}U$ – A new tracer for environmental processes?

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Apart from the production of ^{236}U by fast neutrons in thermonuclear weapons via the reaction $^{238}U(n,3n)^{236}U$, ^{236}U can be also produced in nuclear power plants and fission bombs via $^{235}U(n,\gamma)^{236}U$ using thermal neutrons. In contrast, the principle production path for ^{233}U is via the reaction $^{235}U(n,3n)^{233}U$, which requires fast neutrons with energies above 13 MeV (Gorbatchev et al, 1980). Therefore, an increased production can be expected in thermonuclear weapons using Oralloy (uranium enriched in ^{235}U) as blanket or tamper. Consequently, in average, fallout from nuclear weapons testings should show a higher $^{233}U/^{236}U$ ratio than emissions from thermal nuclear power plants or reprocessing plants which allows source identification for contaminations present in the environment.

However, the cross section of the reaction $^{235}U(n,3n)^{233}U$ for 14 MeV neutrons is only about 0.1 barn, as shown in Figure 1. As there is only little experimental data available for the cross-section of this reaction and the utilization of Oralloy is not readily accessible for all nuclear devices which exploded during the period of atmospheric testing, the ^{233}U fallout from thermonuclear weapons can be only roughly estimated to be around one to two orders of magnitude smaller than ^{236}U fallout. Consequently, neglecting n capture on ^{232}Th in rocks and local contaminations from the ^{232}Th fuel cycle, the environmental concentrations of ^{233}U can be expected to be extremely low, so that its detection is challenging also for the highly sensitive Accelerator Mass Spectrometry (AMS).

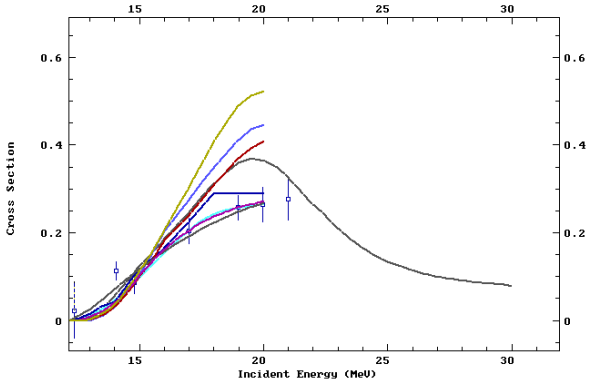

Figure 1. Cross section in barn for the reaction $^{235}U(n,3n)^{233}U$ from model calculations (solid lines) and experimental data (blue squares) (EXFOR/ENDF, 2016)

^{236}U in environmental samples is already routinely analysed at the Vienna Environmental Research Laboratory (VERA) (e.g. Froehlich et al, 2016). VERA has recently increased its detection efficiency such that it is now capable to detect also ^{233}U, which was demonstrated by analysing the concentration of this isotope in different types of environmental sampling material, including Irish Sea sediment, corals from the Pacific Ocean and peat bog samples from Germany. These samples were known to be affected by different contamination sources, i.e. nuclear weapons fallout and the reprocessing plant Sellafield, respectively. In average, the detected $^{233}U/^{236}U$ ratio in the environment is at a level of around 1%. However, the $^{233}U/^{236}U$ ratio in the Irish Sea sediments was around one order of magnitude lower than in the Pacific Ocean corals or the peat bog samples. These findings indicate that the $^{233}U/^{236}U$ can be indeed used for the discrimination between these two contamination sources.

In contrast to Pu, whose isotopic ratios have been successfully used for source identification in the past (Lindahl et al, 2010), uranium shows a considerably higher solubility, and thus, a conservative behaviour in water so that it can be transported over large distances (Sakaguchi et al. 2012). Being isotopes of the same element, the $^{233}U/^{236}U$ ratio is independent from the chemical behaviour in the environment as well as during sample preparation which significantly facilitates the interpretation of the measured data. For these reasons, the $^{233}U/^{236}U$ ratio could serve as a powerful tracer for environmental processes when the principle contamination sources will have been characterized.

After an introduction to possible production paths of ^{233}U and ^{236}U, respectively, first results of the $^{233}U/^{236}U$ ratio detected in samples from the different environmental reservoirs named before will be presented in this talk and the interpretation of the data will be discussed.

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